

Identifying the dominant pairing interaction in heavily electron-doped and single-layer FeSe through Leggett modes

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Heavily electron-doped and single-layer FeSe superconduct at much higher temperatures than bulk FeSe. There have been a number of proposals attempting to explain the origin of the enhanced transition temperature, including the proximity to magnetic, nematic and antiferro-orbital critical points, as well as possible strong interfacial phonon coupling in the case of single-layer FeSe. In this work, we examine the effect of the various mechanisms in an effective two-band model. Within our model, the fluctuations associated with these instabilities contribute to different parts of the effective multiband interactions. We propose to use the collective phase fluctuation between the bands—the Leggett mode—as a tool to identify the dominant effective pairing interaction in these systems. The Leggett mode can be resolved by means of optical probes such as electronic Raman scattering. We point out that the Leggett mode in these systems, if present, shall manifest in the Raman B_{1g} channel.

Introduction – Since its inception in 2008 [1], the unconventional superconductivity in iron-based superconductors (FeSCs) has generated considerable interest. Besides superconductivity, the FeSCs exhibit a rich variety of electronic orders [2–4]. Superconductivity in most FeSC families typically occurs in the vicinity of a stripe magnetic phase. The magnetic order usually follows a nematic transition at a slightly higher temperature [5–7]. The nematic order parameter spontaneously breaks the four-fold rotational symmetry but preserves the translation symmetry of the underlying lattice.

Despite enormous experimental and theoretical progresses, a unified understanding is still lacking regarding the superconducting mechanism in FeSCs [8–12]. The proximity to the magnetic and nematic states indicates strong electron correlations and has led to a number of theories that attribute the superconducting pairing primarily to the magnetic [13–15], orbital [16] and/or nematic fluctuations [17], along with specific predictions for the order parameter symmetry and the gap structure on the multiple bands in the system. However, the complexity originating from the multiple strongly correlated Fe $3d$ -orbitals makes it difficult to unambiguously identify the primary mechanism(s) responsible for the formation of the various electronic orders.

Among all the FeSC compounds, the FeSe family represents a notable outlier. Bulk FeSe superconducts below around 8K at ambient pressure, but T_c increases up to 37K under pressure [18] and can generically reach values above 30K or even higher in heavily electron-doped FeSe. In most cases, the electron-doping is achieved by means of intercalation [19–22], such as in, e.g. $K_x\text{Fe}_{2-y}\text{Se}_2$ and $(\text{Li}_{0.8}\text{Fe}_{0.2})\text{OHFeSe}$. More strikingly, the single-layer FeSe epitaxially grown on SrTiO_3 and BaTiO_3 substrates shows a superconducting transition at temperatures well exceeding 50K [23, 24], with indications of T_c even above 100K [25]. The enhanced superconductivity in these high- T_c FeSe compounds has sparked a substantial series of further investigations.

Although bulk FeSe contains hole and electron Fermi pock-

ets at the Γ - and X/Y -point, respectively, in the Brillouin zone (BZ) similar to other iron-pnictides, but with much smaller size of the Fermi surfaces [26, 27]. Moreover, the hole pocket is absent in the heavily electron-doped [20, 28, 29] and single-layer FeSe [30, 31]. The absence of a hole Fermi pocket at Γ , along with the drastically enhanced T_c , poses a serious challenge to the theories of spin-fluctuation-mediated superconducting pairing based on the quasi-nesting features between the electron and hole pockets [8, 13–15].

Similar to other FeSCs, the undoped FeSe exhibits a transition to nematic order at 90K [32]. However, in strong contrast to the former, the nematic transition is not followed by any long-range magnetic order down to the superconducting transition [33]. Electron doping suppresses the nematic order, while magnetism continues to be absent up to the optimal doping level [34]. Nevertheless, inelastic neutron scattering studies on undoped FeSe reported pronounced spin fluctuations [35], and the standard stripe magnetic order common to other FeSCs does emerge under applied pressure [36–39]. In addition, rich spin excitation spectra are commonly observed in alkali-metal intercalated [40–42] as well as alkali-hydroxy-intercalated [43, 44] electron-doped FeSe-compounds. Furthermore, despite the lack of definitive experimental evidence to date, it is sensible to also pay attention to the antiferro-orbital (AFO) ordering associated with the degenerate and strongly correlated d_{xz} and d_{yz} -orbitals.

The generically enhanced superconducting pairing in the high T_c FeSe compounds seems to connect closely with their peculiar electronic structure with only electron pockets, dichotomy of nematic and magnetic ordering, possible AFO ordering and the presence of their fluctuations, as well as the substrate environment in the case of single-layer FeSe. This naturally motivates the question as to how the nematic, spin and AFO fluctuations may cooperate with the unique electronic structure to strengthen the effective pairing interactions, and how T_c seems to increase further in the presence of interfacial phonon coupling in single-layer FeSe [45, 46].

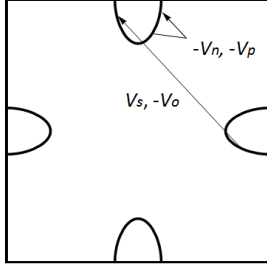


FIG. 1: Sketch of the Fermi surfaces of heavily electron-doped and single-layer FeSe. The primary intra- and interband interactions considered in our model are shown, including the respective SDW-fluctuation and AFO-fluctuation mediated interband interactions V_s and $-V_o$, the nematic-fluctuation induced intraband interaction $-V_n$, and the phonon induced intraband interaction $-V_p$ for single-layer FeSe.

In this work, we do not aim to provide a microscopic theory behind the enhancement of superconductivity as in some recent theoretical works [47–51]. Instead, similar to Li *et al.* [52], we take an effective two-band model and assume a priori the presence of various fluctuation and/or phonon-induced interactions within and between the bands, i.e. intra- and interband interactions. Notably, nematic fluctuations and the interfacial phonon coupling mainly contribute to intraband interaction, while particular types of spin and AFO fluctuations at wavevector (π, π) dominate the interband interaction. When the intraband exceeds the interband interaction, the superconducting state shall exhibit a well-defined collective phase mode – the so-called Leggett mode [53] – which corresponds to the relative phase fluctuations between the two bands. We propose that the existence or nonexistence of the Leggett mode, and the characteristic energy of the Leggett mode if present, can help to elucidate the relative strength of the various contributions to the pairing interactions. The Leggett mode can be probed in optical measurements such as Raman scattering [54, 55]. Note that the Leggett modes in the respective scenarios with dominant intra- and interband interactions have been discussed earlier in a general context [56, 57], while in the present study we focus on heavily-electron doped and single-layer FeSe systems which exhibit distinct Fermi surface geometry.

We also remark that, parallel to the two-band description adopted here, some theories explored the possible crucial role of the incipient hole band at the Γ -point in the BZ. The hole band may develop an incipient Cooper pairing induced by the magnetic fluctuations associated with either the local moments [58] or the itinerant carriers [59]. On this basis, You *et al.* [58] further noted that, since the quasi-nesting between the electron and hole bands is suppressed, superconductivity in high- T_c FeSe systems may have benefited from the absence of a competing itinerant spin-density wave order.

Effective models – We use a two-band model with two electron pockets at the X/Y -points of the single-Fe BZ to mimic the

electronic band structure in heavily electron-doped FeSe, as in Fig.1. The interactions between the low-energy electrons should, in principle, be sensitive to the microscopic details such as the orbital composition at the Fermi level. However, since we are concerned with the properties arising from the couplings between the individual superconducting bands, we may disregard the details of the momentum-space structure of the intra- and interband interactions, but rather take a simplified form for the *integrated* Cooper channel effective interactions within and between the bands.

As in other FeSC's, spin fluctuations are universally present in FeSe compounds. In undoped bulk FeSe, neutron scattering reveals coexisting Néel spin fluctuations at (π, π) and stripe spin fluctuations at $(\pi, 0)$ [35]. Furthermore, heavily electron doped $A_x\text{Fe}_{2-y}\text{Se}_2$ [40–42], and more recently $\text{Li}_{0.8}\text{Fe}_{0.2}\text{ODFeSe}$ [43, 44] were found to exhibit spin resonant excitations at wavevectors surrounding (π, π) . It is of particular interest to ask whether the boosted superconductivity in heavily electron-doped and single-layer FeSe could have benefited from these SDW fluctuations.

The $(\pi, 0)$ fluctuations are ineffective in mediating Cooper pairing in our model due to the lack of a hole pocket at the Γ -point. The fluctuations at wavevector (π, π) (or wavevectors that connect the nearly nested portions of the two Fermi pockets[41, 43], same below), on the other hand, actively scatter electrons between the two pockets and should, therefore, play an important role. Denoting the fluctuating SDW field $\vec{\phi}_s$, the scattering is described by a Yukawa-type coupling between the SDW field and the carriers in the effective action,

$$\mathcal{S}_s = \lambda_s \int d\vec{q} \vec{\phi}_s(\vec{q}) \cdot \sum_{\alpha, \beta} \int d\vec{k} c_{\vec{k}+\vec{q}, \alpha}^\dagger \vec{\sigma}_{\alpha\beta} c_{\vec{k}, \beta} + h.c. \quad (1)$$

Integrating out the SDW field returns a spin-dependent effective interaction peaking at momentum transfer (π, π) , i.e. $V_s(\vec{k}, \vec{p}) \propto -\chi_s(\vec{k} - \vec{p}) \vec{\sigma}_{\alpha\beta} \cdot \vec{\sigma}_{\gamma\delta}$, where χ_s stands for the SDW magnetic susceptibility. This wavevector connects the two Fermi pockets, hence the interaction is predominantly interband. Such an effective interaction in the singlet pairing channel amounts to a repulsive interaction peaking at the same momentum transfer, thereby promoting sign-changing superconducting gaps on the two bands, i.e. a node-less d -wave pairing.

Likewise, AFO fluctuations in FeSCs may also develop predominantly at wavevectors (π, π) and $(\pi, 0)$. We consider the former wavevector, for which the fluctuations scatter electrons between the two pockets. There is, however, an important distinction from the SDW fluctuations, in that here the scattering is spin-independent,

$$\mathcal{S}_o = \lambda_o \int d\vec{q} \phi_o(\vec{q}) \sum_{\sigma} \int d\vec{k} c_{\vec{k}+\vec{q}, \sigma}^\dagger c_{\vec{k}, \sigma} + h.c. \quad (2)$$

This leads to an effective interaction in the Cooper channel, $V_o(\vec{k}, \vec{p}) \propto -\chi_o(\vec{k} - \vec{p})$, which is primarily attractive and interband, thus favoring a sign-conserving s -wave pairing. As

a consequence, the AFO fluctuation and the SDW fluctuation mentioned above compete against each other.

The nematic susceptibility χ_n , whether spin- or orbital-driven, peaks at zero momentum. Hence nematic fluctuations are effective in scattering electrons by small momenta, in a manner given by the effective action,

$$\mathcal{S}_n = \lambda_n \int \vec{q} \phi_n(\vec{q}) \sum_{\sigma} \int d\vec{k} c_{\vec{k}+\vec{q},\sigma}^{\dagger} c_{\vec{k},\sigma} + h.c. \quad (3)$$

This scattering is also spin-independent. The induced effective electron interaction, $V_n(\vec{k}, \vec{p}) \propto -\chi_n(\vec{k} - \vec{p})$, is attractive and predominantly intraband. This interaction alone drives electron pairing, and should give rise to degenerate sign-changing (*d*-wave) and sign-conserving (*s*-wave) gaps. The degeneracy can be lifted by either interband interactions or interband hybridization, the latter of which has been discussed in Ref. 60.

Taking together, the primary multiband interactions can be expressed as,

$$\hat{V} = \begin{pmatrix} -V_n & V_s - V_o \\ V_s - V_o & -V_n \end{pmatrix}, \quad (4)$$

where we take $V_n, V_s, V_o > 0$ for notational clarity. Solving a coupled BCS gap equation using (4) yields gap functions on the two bands, the more attractive of which characterizes the stable ground state. Since the two bands have the same density of states, the gap functions are equivalent to the eigenvectors of \hat{V} . The solution thus obtained denotes the relative sign and magnitude of the band gaps. The preference between sign-changing and sign-conserving pairings is determined by relative strength of the two interband interactions, i.e. the former is favored if $V_s > V_o$, otherwise the latter is more stable.

A cooperative mechanism of a certain subset of the multiband interactions may be crucial for the boost of T_c . In particular, the intra- and net interband interactions do not compete: since the most negative eigenvalue of \hat{V} is given by $-V_n - |V_s - V_o|$, both intra- and interband interactions act to strengthen pairing. The enhancement is most effective when either V_s or V_o dominates the interband interaction.

In the proximity of the SDW (nematic, AFO) quantum critical point, the interaction V_s (V_n, V_o) is substantially enhanced and may dominate the interactions. This naturally leads to much enhanced superconductivity, provided that the normal state carriers remain sufficiently coherent quasiparticles. The aspect of the loss of quasiparticle coherence due to the quantum fluctuations near the critical point is, however, beyond the scope of our theory.

In light of the striking observation of a replica band in single-layer FeSe suggestive of a strong small-momentum scattering by the interfacial phonons [46], phonon-induced effects should be properly accounted for in this system. This gives an effective action analogous to the one formulated for nematic fluctuations (3). As a consequence, the phonon coupling gives rise to an attractive interaction $-V_p$ ($V_p > 0$) in

the same fashion as the nematic fluctuations mediating $-V_n$. Hence the total effective interaction becomes,

$$\hat{V} = \begin{pmatrix} -V_n - V_p & V_s - V_o \\ V_s - V_o & -V_n - V_p \end{pmatrix}. \quad (5)$$

The effective pairing interaction is then given by $-V_n - V_p - |V_s - V_o|$, from which it is easy to see the conducive role of the interfacial phonons in enhancing superconductivity.

In the following, we first discuss the existence/nonexistence of Leggett mode in the presence of various dominant pairing interactions, and then proceed to discuss the detection of the Leggett mode when it does exist.

Leggett mode energy – As was originally conjectured by Leggett [53], the interband interaction gives rise to an effective Josephson coupling between the superconducting order parameters on different bands, which locks their relative $U(1)$ phase. Under external perturbations, the relative phase can oscillate in time, costing a finite amount of energy that is determined by the interband coupling. This is the *Leggett mode*. In essence, this collective mode corresponds to a fluctuation between the leading and subleading superconducting states [61]. In this regard, the Leggett mode shares the same spirit as, and thereby constitutes a special example of, the so-called Bardasis-Schrieffer (BS) mode [62], which has been discussed previously in the context of iron-pnictide superconductors [63–66]. Note that the BS mode can also exist in single-band systems.

Naturally, the characteristic energy of the Leggett mode encodes crucial information about the multiband interactions in FeSe systems. Below we analyze the Leggett modes in the presence of various configurations of multiband interactions. The expressions of the Leggett mode energy is derived in Ref. 69, and we shall directly quote results therein.

We first ignore the phonon contribution V_p . Of particular interest are the two limiting cases where the pairing is driven primarily by the nematic or by SDW/AFO fluctuations. In the former, $V_n \gg |V_s - V_o|$, the interband Josephson coupling reads $J = |V_s - V_o|/(V_n^2 - |V_s - V_o|^2) > 0$ (see Ref. 69). Both solutions to the gap equation correspond to attractive superconducting channels, i.e., leading *d*-wave with subleading *s*-wave pairing if $V_s > V_o$, or vice versa if $V_s < V_o$. Thus, a coherent Leggett mode exists, and its resonance energy is given by,

$$w_L = \sqrt{2 \frac{J}{N_0}} \Delta_0 = \sqrt{\frac{2}{\lambda}} \sqrt{\frac{|V_s - V_o|}{V_n - |V_s - V_o|}} \Delta_0, \quad (6)$$

where Δ_0 is the superconducting gap, N_0 is the density of states of a single band, and $\lambda = N_0(V_n + |V_s - V_o|)$ gives the effective coupling strength in the leading superconducting channel. Taking a rough approximation $\lambda \sim 1$, $w_L \sim \sqrt{2|V_s - V_o|/V_n} \Delta_0 \ll 2\Delta_0$, which is much smaller than the quasiparticle continuum edge at $w = 2\Delta_0$. Such a soft mode reflects the near-degeneracy between the leading and subleading pairing states, and equivalently the relative ease in fluctuation

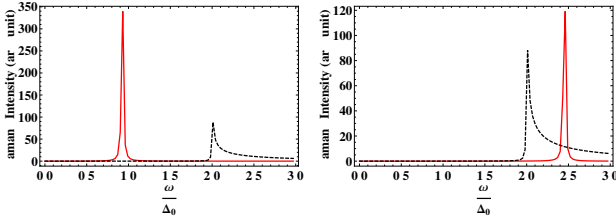


FIG. 2: Schematic Raman intensity in the B_{1g} channel for two models with the ratio of inter- and intraband interactions 0.3 (left) and 0.75 (right). In both cases the intraband interaction is stronger than the interband one. The continuum contribution (black dashed) shows a peak at $w = 2\Delta_0$, while the Leggett resonance (red solid) occurs at w_L . We have assumed an isotropic superconducting gap on the two bands, taking the approximation $\lambda \sim 1$ in (6), and used a small imaginary component $\tau = 0.002\Delta_0$ to yield broadened peaks. These calculations follow the formulation explained in a previous work [67]. On the right figure we have ignored the quasiparticle damping of the Leggett resonance.

tuating the relative phase between the two bands when the interband interaction is weak.

In the other limit, $V_n \ll |V_s - V_o|$, according to Ref. 69, no coherent Leggett mode is present. A simple explanation is as follows. When interband interaction dominates, the interaction \hat{V} yields one attractive and one repulsive solution in the superconducting channel, with eigenvalues $-V_n - |V_s - V_o|$ and $-V_n + |V_s - V_o|$. Since the repulsive channel does not represent a true superconducting instability, no subleading superconducting channel exists to be excited to. Thus, no coherent Leggett mode appears under these conditions.

In the intermediate regime with $|V_s - V_o| \lesssim V_n$, w_L is again given by (6). The characteristic energy w_L increases with growing $|V_s - V_o|/V_n$, exceeding the continuum edge for $|V_s - V_o| \sim V_n/2$. In this regard w_L is a qualitative measure of the relative strength between the intra- and interband interactions. Finally, in accordance with the discussions in the previous section, the phonons strengthen intraband interactions. Therefore, the addition of V_p broadens the parameter range where a coherent Leggett mode can appear.

Detection of the Leggett mode – The Leggett modes couple to electromagnetic fields and, hence, can be excited by photons in optical measurements, such as the electronic Raman scattering, as has been demonstrated for the two-band superconductor MgB_2 [55]. They manifest as resonance features in the Raman spectrum when the frequency of the incident photons matches that of the Leggett modes in an appropriate scattering channel. Note when w_L exceeds the continuum edge, the Leggett mode becomes damped by quasiparticle excitations, and the broadened resonance peak overlaps with the quasiparticle continuum spectrum at $w > 2|\Delta|$, wherein the measured Q -factor may be small (as is the case in MgB_2 [55]).

In usual cases, such as in MgB_2 [55], the leading and subleading superconducting states belong to the same irreducible point group representation, i.e. the relative $U(1)$ phase os-

cillation does not alter the pairing symmetry. Hence the resonance associated with the Leggett mode only arises in the Raman A_{1g} channel. However, in heavily electron-doped and single-layer FeSe, the phase oscillation amounts to a fluctuation between the s - and d -wave states, which changes the Cooper pair angular momentum by $2\hbar$. This originates from the unique Fermi surface topology with the two Fermi pockets locating around the X and Y -points in the BZ (Fig.1). Consequently, the Leggett mode under consideration will couple only to the Raman B_{1g} channel, and the resonance will emerge in this channel, as was also noted in a recent work [66] cast in more general context. Figure 2 shows two representative B_{1g} -channel Raman spectra when the intraband interaction plays the leading role in driving the pairing.

Highly relevant Raman scattering measurements have been performed on a heavily electron-doped intercalated compound, $\text{Rb}_{0.8}\text{Fe}_{1.6}\text{Se}_2$ [68]. There, below the continuum edge, apart from a phonon mode, no additional peak was clearly visible in the B_{1g} channel with features that could be associated with a Leggett resonance. It is also unclear whether any resonance is present in the B_{1g} spectrum above the continuum edge. Hence we cannot conclude on the qualitative comparison between the intra- and interband interactions. Nonetheless, following our arguments above, the absence of such a resonance below $2\Delta_0$ suggests that the interband interaction at least constitutes a non-negligible ingredient of the total effective pairing strength in this particular compound. Corroborating the significance of interband interactions, the superconducting magnetic resonant modes observed in neutron scattering in several heavily electron-doped intercalated compounds [40–42], including $\text{Rb}_{0.8}\text{Fe}_{1.6}\text{Se}_2$ [41], appeared at wavevectors which most likely connect the two bands [41, 43].

Assisted by the strong coupling to interfacial phonons [46], the intraband interaction is expected to be boosted significantly in the single-layer FeSe grown on ATiO_3 substrates. A coherent Leggett mode is thus more likely to emerge in these systems. However, due to the finite optical penetration into the substrate, the Raman spectroscopy may see a much stronger background noise signal, making it difficult to disentangle the authentic response of the FeSe layer. It is, thus, necessary to devise a careful Raman measurement to search for such a Leggett resonance there.

Conclusions – In this work, we outlined the possible main sources of the multiband interactions in the two-band high- T_c heavily electron-doped and single-layer FeSe superconductors. The nematic fluctuations and/or interfacial phonons contribute primarily to the intraband interaction, while the SDW and AFO fluctuations at momentum (π, π) (or similar wavevectors connecting the two pockets) mainly drive competing interband interactions. If the net interband interaction is weaker than the intraband one, a novel collective phase excitation—a Leggett mode—shall arise. We proposed that optical probes such as Raman spectroscopy can provide crucial information regarding the relative strength of the various contributions to the effective pairing glue.

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Supplemental Material for **“Identifying the dominant pairing interaction in heavily electron-doped and single-layer FeSe through Leggett modes”**

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Here, we derive the characteristic energy of the Leggett modes in our effective two-band model introduced in the main text. We take the effective interaction (4) in the main text for illustration. For simplicity, we include below only the nematic and spin fluctuation mediated interactions, V_n and V_s . The other interactions can be accounted for easily in an analogous way. The coupled BCS gap equation is written as,

$$\begin{pmatrix} \Delta_1 \\ \Delta_2 \end{pmatrix} = -N_0 \ln \left(\frac{\pi W_c}{2T_c} \right) \begin{pmatrix} -V_n & V_s \\ V_s & -V_n \end{pmatrix} \begin{pmatrix} \Delta_1 \\ \Delta_2 \end{pmatrix}, \quad (1)$$

where N_0 is the density of states of a single band, T_c is the superconducting transition temperature, and W_c is some characteristic cutoff energy. In principle, the cutoff should be different for interactions mediated by nematic and magnetic fluctuations, but for simplicity we take it to be the same for both. Solving the gap equation is equivalent to diagonalizing \hat{V} , the latter of which returns the effective pairing interaction in the eigen-channels: $-V_n - V_s$ and $-V_n + V_s$. The two eigenvectors are $(1, -1)$ and $(1, 1)$. The first solution corresponds to the superconducting ground state, which describes a sign-changing node-less d -wave pairing. Note if the attractive interband interaction $-V_o$ is included and if $V_o > V_s$, the second solution, i.e. the sign-preserving s -wave state, is favored.

Following the procedures formulated elsewhere [1–3], the effective Josephson coupling between the bands can be captured in the following effective action,

$$S_\Delta = \int \hat{\Delta}^\dagger (-\hat{V}^{-1}) \hat{\Delta} - \sum_{l=1,2} \text{Tr} \ln G_l^{-1}, \quad (2)$$

where $\hat{\Delta} = (\Delta_1, \Delta_2)^T$ denotes the superconducting order parameter on the two bands, and the l -band Gor'kov Green's function is given by

$$\hat{G}_l^{-1} = - \begin{pmatrix} \partial_\tau - \frac{\nabla^2}{2m} - \mu & -\Delta_l \\ -\Delta_l^* & \partial_\tau + \frac{\nabla^2}{2m} + \mu \end{pmatrix}. \quad (3)$$

In (2), we have assumed that \hat{V}^{-1} is non-singular, i.e. $\det \hat{V}$ is non-vanishing. The interband Josephson coupling J is determined by the off-diagonal element of the inverse of $-\hat{V}$,

$$J = \frac{V_s}{\det \hat{V}}, \quad (4)$$

Considering now small deviations of the $U(1)$ phase of the order parameter from the stable state, $\theta_l = \theta_{0l} + \phi_l$ ($l = 1, 2$), the action in Eq.(2) can be expanded with respect to the ϕ_l 's as,

$$S_\Delta[\phi] = \int \vec{q} \sum_{w_n} \hat{\phi}^T(-w_n, -\vec{q}) \mathcal{M} \hat{\phi}(w_n, \vec{q}), \quad (5)$$

where $\hat{\phi}(w_n, \vec{q}) = (\phi_1, \phi_2)^T(w_n, \vec{q})$, and the matrix,

$$\mathcal{M} = \begin{pmatrix} \mathcal{K} - J\epsilon_{12} & J\epsilon_{12} \\ J\epsilon_{12} & \mathcal{K} - J\epsilon_{12} \end{pmatrix} \quad (6)$$

where $\mathcal{K} = N_0(w_n^2 + \bar{v}_F^2 q^2/2)$ with N_0 being the density of states on a single band and \bar{v}_F being the average Fermi velocity on the bands, and $\epsilon_{12} = \cos(\theta_{01} - \theta_{02})|\Delta|^2$. Since the repulsive V_s favors sign-changing pairing, $\epsilon_{12} \equiv -|\Delta|^2$ in our model. After an analytic continuation by replacing $iw_n \rightarrow w + i0^+$, the dispersion relations for the phase modes may be obtained by diagonalization (6),

$$w_G^2 = \frac{1}{2} \bar{v}_F^2 q^2, \quad (7)$$

$$w_L^2 = 2 \frac{|\Delta|^2}{N_0} J + \frac{1}{2} \bar{v}_F^2 q^2. \quad (8)$$

Here w_G denotes the usual gapless $U(1)$ Goldstone mode, which would be massive had we properly included the vector potential in our formalism; w_L is the Leggett mode, whose excitation gap is determined by interband Josephson coupling. Note that if $J < 0$, as would be the case for $V_s > V_n$, no physical solution exists for w_L , i.e. the Leggett mode is overdamped. This peculiar scenario corresponds to the absence of coherent relative phase oscillations when the pairing is overly dominated by the interband interaction [4]. To understand this, first note that the Leggett mode corresponds essentially to a fluctuation between the leading and subleading pairing channels mentioned above (d - and s -waves in our case). If the subleading channel becomes repulsive, i.e. $-V_n + V_s > 0$, there exists no true subdominant superconducting channel for the ground state to coherently excite to. As a consequence, a coherent Leggett mode cannot exist in this scenario.



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